

PATENT SPECIFICATION

1,062,307

1,062,307



Inventors: LESLIE ALFRED MUIRHEAD, EDWARD LAWRENCE HILL and BRIAN WILFRED FORSTER.

Date of filing Complete Specification: February 7, 1966.

Application Date: March 17, 1965.

No. 11311/65

Complete Specification Published. March 22, 1967

© Crown Copyright 1967.

Index at Acceptance:—C3 C (1B2, 2); B5 A (1G4X, 1G10, 2A1, 2A3, 2B1, 2B2); C3 P (7D2A1, 7F1, 7F2).

Int. Cl.:—C 08 j 1/26 // B29d.

COMPLETE SPECIFICATION

DRAWINGS ATTACHED

Improvements in and relating to the Manufacture of Particulate Expandable Polymers and to the Manufacture of Expanded Polymers Therefrom

We, SHELL INTERNATIONALE RESEARCH MAATSCHAPPIJ N.V., a company organised under the laws of The Netherlands, of 30 Carel van Bylandtlaan, The Hague, The Netherlands, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

10 The present invention is concerned with the manufacture of expandable particles of a polymer of a vinyl-aromatic compound and for a vinylidene-aromatic compound (hereinafter referred to as a thermoplastic organic polymer), for example, polystyrene the term 15 polymer including a copolymer or a polymer composition; with the resulting expandable particles; and with the manufacture of an expanded (i.e. a foamed) polymer of cellular form, including shaped articles, therefrom.

It is known that a thermoplastic organic polymer such as polystyrene can be expanded to a cellular form by intimately incorporating a vaporisable expanding agent 25 in the polymer and then heating the polymer under conditions permitting the expanding agent to vaporise to form a larger number of individual, enclosed spaces or cells within the polymer. In the case of polystyrene it is 30 common to use as the vaporisable expanding agent a normally-liquid, vaporisable organic substance, such as technical pentane, which when intimately incorporated in the polymer forms an "expandable polymer." 35 On heating such an expandable polymer, the expanding agent vaporises in a large number of individual locations within the polymer to form said cells. On cooling, the resulting polymer is of cellular form: such a polymer 40 is referred to herein as an "expanded polymer."

It is also known to manufacture shaped, cellular articles from polymers by forming

an expandable polymer in particulate form, i.e. expandable polymer particles, and then 45 heating a mass of "pre-expanded" expandable particles in an enclosed mould of appropriate shape. The expandable polymer particles are normally "pre-expanded", that is they are heated to effect at least a major 50 part of the expansion of which they are capable, prior to charging the particles to the mould so that formation of the shaped cellular article is in fact effected by the fusion together of the pre-expanded particles in the 55 mould with only a minor amount of further expansion occurring therein. An early description of such technique occurs in U.K. Patent Specification No. 715,100. To avoid the necessity for transporting the more bulky 60 pre-expanded particles from the manufacturer of expandable particles to the user thereof, pre-expansion of expandable particles is usually carried out by the user, i.e. the manufacturer of shaped cellular polymer 65 articles, and such pre-expansion is usually effected by contacting the expandable polymer particles with live steam.

For many years expandable polystyrene particles have been manufactured by incorporating a vaporisable expanding agent in 70 polystyrene beads obtained by a suspension polymerisation process. More recently it has also been proposed to incorporate such an expanding agent in heat-plastified polystyrene in an extruder and to form expandable particles by cutting the expandable, 75 filamentary extrudate. Since such an extrudate is an expandable material at an elevated temperature the prior proposals have included special steps designed to prevent any 80 substantial premature expansion of the extrudate, such steps comprising immediately contacting the extrudate with a liquid coolant such as water to cool the extrudate to 85 a temperature below 50°C or extruding the

BEST AVAILABLE COPY

expandable polymer into a pressure zone in which the extrudate remains until it has been cooled to a temperature at which it is incapable of expanding to any significant extent.

5 The use of a pressure zone brings with it operational complications, particularly when the extrudate is chopped into particles within the pressure zone; whilst sudden cooling of the extrudate to below 50°C is said to

10 "freeze" the polymer in an oriented or strained condition and in such case a subsequent annealing operation involving heat treatment at a temperature between 50 and 75°C has been proposed as a means of relaxing the orientation or strain existing within the expandable polymer particles.

According to the present invention a process for the manufacture of expandable particles of a thermoplastic organic polymer (as hereinbefore defined) in which a vaporisable expanding agent is incorporated in a heat-plastified mass of said polymer and the heat-plastified polymer is extruded in filamentary form and cut into particles, which comprises extruding the heat-plastified polymer, containing said expanding agent, into an essentially gaseous environment at atmospheric or substantially atmospheric pressure and immediately cooling the extruded polymer by contact with a fluid at a temperature between 60 and 80°C in such a manner that the temperature of the extruded polymer does not fall below 60°C, maintaining the temperature of the extruded polymer between 60 and 80°C during a normalising period (as herein defined) immediately following extrusion thereof and before and/or after cutting the extruded polymer into particles, and thereafter cooling the polymer particles to ambient temperature. Preferably the extruded polymer is cut into particles and the resulting particles are then subjected to normalising conditions, although it is also possible to normalise the polymer in filamentary form prior to cutting or for the normalising period to extend both before and after the cutting operation.

The present invention also includes the resulting particulate expandable polymer i.e. expandable particles of a thermoplastic organic polymer; and expanded particles and shaped articles formed therefrom by any known or suitable expansion technique or moulding process.

55 The reference herein to an essentially gaseous environment means an environment which is either wholly gaseous or mainly gaseous to the extent that liquid droplets, for example, a water spray can also be present.

60 Said normalising period is defined herein as the minimum period, i.e. length of time, for which an extruded expandable polymer in filamentary or particulate form must be maintained at a temperature within the range 65 60 to 80°C and at normal pressure prior to

being cooled to ambient temperature in order that the size of at least 90% of the cells of an expanded particle, obtained by freely expanding such an expandable particle (or an expandable particle cut from such expandable filamentary polymer) by heating under conditions resulting in a maximum increase in volume of the particle, is not greater than 100 microns (in diameter). In general a cell size of about 100 microns is the maximum cell size currently acceptable commercially in the manufacture of cellular polystyrene articles by moulding expanded polystyrene particles.

The process of the present invention has the operational advantage of producing normalised expandable polymer particles which do not require any subsequent annealing operation and since the temperature of the extruded polymer is not allowed to fall below 60°C until normalisation has been achieved, it is possible that one avoids the intermediate stage in which, according to the prior art, orientation or strain is produced. It has been found that operation in accordance with the present invention permits the attainment of expandable polystyrene particles which, from the point of view of their end-use behaviour in a subsequent moulding operation leading to shaped cellular polystyrene articles, closely resemble the well-known expandable polystyrene particles obtained by incorporating an expanding agent in the particulate product of a suspension polymerisation process.

One of the major difficulties which can arise in the manufacture of expandable polymer particles by methods involving incorporating a vaporisable expanding agent in the heat-plastified polymer is the problem of cell size in the resulting expanded polymer and, whatever the mechanism of the normalising treatment of the present invention may be, it has been found that operation in accordance with the present invention enables a satisfactory cell size (i.e. 90% below 100 microns) to be attained in the ultimate cellular polymer.

In carrying out the present invention it is essential to normalise the extruded expandable polymer either as particles or in filamentary form prior to cutting into particles. The extrudate comprising a thermoplastic organic polymer, for example, polystyrene in a heat-plastified form and containing a vaporisable expanding agent dispersed therein, can be cooled to and normalised at 60-80°C whilst in the form of a filament (or a plurality of filaments). Such a normalised filament can then be cut into particles whilst said filament is at 60-80°C or after first cooling to ambient temperature; but preferably said filamentary extrudate is cut into particles immediately following extrusion thereof by cutting the extrudate at the extrusion ori-

face of the extruder (face-cutting) or shortly after extrusion so that the extruded expandable polymer is normalised in particulate form. Operation in this preferred manner has practical advantages as compared to normalisation in filamentary form. When employing the preferred technique the extruded filament is immediately contacted, in an atmospheric or substantially atmospheric pressure environment, with a fluid at a temperature between 60 and 80°C whilst simultaneously, or within a short time of extrusion, cutting said extruded filament into particles, and thereafter contacting the resulting particles with a fluid at a temperature between 60 and 80°C under conditions such that the temperature of both said extruded filament and the resulting particles does not fall below 60°C during the required normalising period immediately following extrusion.

The extruded filament can be cooled by contact with a liquid or gas at a temperature of 60-80°C, but preferably with a liquid, e.g. water. When face-cutting is employed the liquid is preferably in droplet form, the expandable polymer being sprayed therewith as it is being extruded. When the filamentary extrudate is cut into particles subsequent to cooling with a liquid at 60-80°C, the extrudate can be passed immediately into a body of such liquid disposed as near as possible to the extrusion orifice(s) and if desired the extrudate can also be sprayed with liquid prior to entering the body of liquid. The normalising of the extruded filament or particles cut therefrom is conveniently achieved by contacting said filament or particles with a body of cooling liquid at 60-80°C, which is preferably water. Cooling of the normalised filament(s) or particles to ambient temperature is conveniently achieved by contacting said filament(s) or particles with air and this can be a combined cooling and drying operation to remove liquid, e.g. water, physically associated therewith.

Whilst the temperature of the heat-plastified polymer as it is being extruded (i.e. its extrusion temperature at the die-head) will depend, *inter alia*, on the softening temperature of the polymer (which may be lower than that of the polymer *per se* due to the presence therein of the vaporisable expanding agent), the extrusion temperature should always be high enough to avoid the possibility of solidification of the polymer in the extrusion orifice(s) of the die-head. The required extrusion temperature will usually depend on the dimensions, and in particular the cross-sectional area, of the extrusion orifice; and on the extent to which the die-head itself is cooled by the cooling fluid at 60-80°C with which the extrudate is contacted, particularly when using face-cutting technique. For example, for polystyrene con-

taining 4 to 8% by weight of technical pentane as the expanding agent, the extrusion temperature of the heat-plastified polymer can be as low as between 105 and 115°C, e.g. about 110°C, if the extrusion orifice(s) is (are) of the order of 2 to 3 millimetres in diameter and if there is no significant heat loss from the heat-plastified polystyrene in the die-head, but higher temperatures should be employed when extrusion orifices of smaller diameter are employed. In general extrusion temperatures for polystyrene containing technical pentane can be up to 150 to 160°C depending on the extrusion conditions employed. Temperatures of 130 to 160°C are advantageous when using small diameter extrusion orifices i.e. below 1.0 millimetres, e.g. of the order of 0.5 to 0.75 millimetres, in diameter.

Normalising can be carried out by contacting the extrudate with said fluid at a temperature between 60 and 80°C e.g. about 70°C for a period, e.g. between 30 and 60 minutes for expandable polystyrene particles, depending on the physical form, e.g. the particle size, of the extrudate. A normalising liquid, for example, water or brine or other aqueous media, e.g. mixtures of water and glycerol or ethylene glycol, can be used, and when the extrudate is normalised in particulate form the particles are subsequently separated from the normalising liquid in any convenient manner, e.g. by employing a screen which can be an inclined, vibratable screen or a perforated travelling belt. When a normalising liquid such as water is used the particles are thereafter dried and cooled to ambient temperature e.g. in a current of air.

It will be understood that a plurality, e.g. between 20 and 50, of filaments of expandable polymer will usually be extruded simultaneously using a multi-orifice die-head when carrying out the present invention on a manufacturing scale. It is usual to employ circular extrusion orifices with the result that the filamentary extrudate is of essentially circular cross-section: whilst such a filament is of rod-like form it is usual in the art to refer to it as a strand and the term "strand" will be used in this general sense in the following specific description.

Said thermoplastic polymer can be polystyrene or other polyvinyl-aromatic compound and/or polyvinylidene-aromatic compound or a copolymer of a vinyl-aromatic compound and/or vinylidene-aromatic compound e.g. with acrylonitrile, methyl methacrylate, acrylic acid or methacrylic acid; although the present invention is particularly applicable to polystyrene.

Said vaporisable expanding agent can be a normally liquid organic substance having substantially no solvent action on said thermoplastic polymer or a mixture of such

organic substances; and if desired such a substance or mixture thereof can be used together with a small amount of a solvent for said polymer. Preferably said expanding agent is an organic liquid boiling, at normal pressure, below 95°C and usually below about 80°C, e.g. a hydrocarbon or mixture thereof such for example as n-pentane or technical pentane or a mixture of n-pentane and iso-pentane containing between 30 and 80% by weight of iso-pentane. If desired said polymer can contain an additive, e.g. a low molecular weight polyethylene wax, advantageously in an amount which is between 0.05 and 0.1% by weight of the polymer, as referred to in the specification of our co-pending U.K. Patent Application No. 146/62 Serial No. 997356.

The process of the present invention can be carried out either by cutting the expandable polymer as it is extruded from the die orifice(s) using any known or suitable face-cutting technique or by extruding the polymer in strand form and subsequently chopping or cutting the strand(s) into particles using any known or suitable strand-cutting technique. In each case the expandable polymer is immediately contacted with cooling fluid at 60 to 80°C, either as it issues from the die orifice(s) as when face-cutting or as soon as practicable thereafter when a strand cutting technique is employed. When strand-cutting is used it is preferable to carry out the cutting operation before or at least during the initial stage of the normalising period in order to avoid handling long strands: and preferably the degree of draw-down of the extruded strand(s) is as small as possible to reduce the risk of strand breakage and in general the degree of draw-down should not exceed that which will normally occur in a strand-cutting operation employing the minimum haul-off tension in the strands. When employing the strand-cutting technique it is usually unnecessary to direct cooling fluid against the face of the die-head providing the extruded strands are substantially immediately contacted with said cooling fluid so as to reduce the temperature of at least the surfaces of the strands to the normalising temperature of 60 to 80°C as quickly as possible. However, it is possible to direct cooling fluid, e.g. a spray of liquid droplets, against the face of the die-head if desired and operation in this way is recommended if the extrudate tends to expand prematurely prior to entering for example a body of cooling liquid at 60 to 80°C disposed adjacent the die-head. In general, however, when using water at 60 to 80°C as the cooling fluid, no cooling of the die-head itself will occur when employing a strand-cutting technique, whereas when employing a face-cutting technique it has been found advisable to ensure that the cutting

face is kept "wetted" with a cooling liquid in order to prevent smearing of the polymer on the cutting face, which impairs the face-cutting operation. The removal of heat from the die-head as the result of cooling liquid contacting the die face could cause premature solidification of polymer in the die-head. However, this can be prevented in several ways including, for example, supplying heat to the die-head itself by providing it with an electrical heating element or passage for the circulation therethrough of a heating fluid e.g. steam, preferably together with the provision of thermal insulation between the die face and the body of the die-head as will be described later in detail in connection with the use of face-cutting technique in carrying out the present invention. Alternatively or in addition one can operate at higher extrusion temperatures, e.g. for polystyrene between 130 and 160°C, providing, of course, the extrusion temperature is not so high as to cause undesirable expansion of the expandable polymer as it issues from the die-head. Whilst such measures are considered to be more important in the case of face-cutting, it is to be understood that the relationship between polymer solidification and extrusion orifice dimensions referred to previously can render such measures applicable to strand cutting as well.

Whether face-cutting or strand-cutting techniques are employed, the vaporisable expanding agent is incorporated in the polymer in a screw extruder comprising a barrel providing a heat-plastifying section followed by a dispersion section to which the expanding agent is supplied and in which it is incorporated in the heat-plastified polymer fed by the extruder screw from the first section. Heat is supplied to the polymer in the first section, e.g. electrically or by means of a heat-transfer fluid circulated through a jacket surrounding the first section; and usually heat is removed from the polymer in the second section by means of a heat-transfer fluid e.g. water circulated through a jacket surrounding the second section. The extruder screw extends through the length of both sections of the extruder and is profiled to provide for agitation and forward feeding of the polymer in the first section and the exertion of extrusion pressure thereon; and for mixing of the heat-plastified polymer and the expanding agent in the second section. The second section is followed by the die-head through which the expandable polymer formed in the second section is extruded under pressure exerted on the polymer in the first section, the degree of heat removal in the second section being controlled in accordance with the desired extrusion temperature.

A convenient way of carrying out the pre-

sent invention employing a strand cutting technique for manufacturing e.g. expandable polystyrene particles is illustrated diagrammatically in Figure I of the accompanying drawings. Referring to Figure I, polystyrene granules are fed to a screw extruder 10 having a heating jacket 11 extending over part of its length in which the extruder screw 12 is of conventional profile for heat-plastifying the polystyrene and advancing the heat-plastified polymer under extrusion pressure towards the extruder die-head 13. The remaining part 14 of the extruder 10 constitutes a dispersion section in which technical pentane supplied through a line 15 is incorporated in the heat-plastified polystyrene. A cooling jacket 16 surrounds the dispersion section and that part 17 of the extruder screw lying within the dispersion section is shaped to provide the agitation required to effect an intimate dispersion of the pentane in the polystyrene. The pentane-containing polystyrene is extruded as strands 18 from the die-head 14 (four such strands being shown) and these strands 18 are immediately led into a bath 19 containing water at between 60°C and 80°C to effect cooling of at least the skin of the strands 18 to the temperature of the water before cutting the strands into particles using a conventional strand cutter 20. The resulting particles 21 fall into a slurry tank 22 also containing water at between 60°C and 80°C which constitutes a reservoir for the subsequent normalising stage which is carried out in a tower 23 comprising an internal agitator 24 shaped in such a manner as to progressively lift up through the tower 23 the particles supplied to the base of the tower as an aqueous slurry (suspension), via a line 25, a pump 26 and a line 27, from the slurry tank 22. The tower 23 also contains an internal heating coil 28 for maintaining the water in the tower 23 at a temperature between 60 and 80°C. The size of the tower 23 in relation to the average residence time of the particles 21 in the slurry tank 22 and the particle throughput rate in the tower 23 is such that the particles formed by the cutter 20 are subjected to the required normalisation i.e. the particles are maintained at a temperature between 60 and 80°C for a normalising period (e.g. 40 minutes) prior to discharge from the top of the tower 23. The normalised particles are discharged from the tower 23 through a line 29 onto the lower end of a conveyor 30 having a perforated belt to permit water to drain from the particles into a reservoir 31 in which the water is heated for return via line 32 to the slurry tank 22. The particles 21 are conveyed to a bin 33, if desired via an air dryer shown in dotted lines at 34.

In the alternative way of carrying out the process of the present invention in which the

extruded polymer containing a vapourisable, normally liquid expanding agent e.g. technical pentane, is extruded and face-cut at the die-head, the face-cutting is preferably effected by means of a rotatable cutter whilst the face of the die-head is simultaneously contacted with droplets of water at a temperature of 60 to 80°C sprayed onto said face. The water droplets at this temperature are conveniently formed by spraying wet steam towards the face of the die-head. The resulting particles of expanded polymer are washed by the water spray into a body of water maintained at a temperature of 60 to 80°C, e.g. about 70°C, in which the particles remain for the desired normalising period of e.g. 30 to 60 minutes. The die-head is provided with means for counteracting the heat loss which occurs as the result of the water spray on the die face and a preferred way of achieving this is to employ a die-head of sandwich construction which comprises a front plate constituting the die face against which the cutter rotates, a back plate which is secured to the extrusion end of the extruder and, between the front and back plates, an insulating plate to provide thermal insulation between the die face and the extruder. The back plate preferably also functions as a heating plate for which purpose it is preferably provided with a passage or passages for the circulation of a heating fluid: alternatively the back plate can embody or be surrounded by an electrical heater, but heating by means of a circulating heating fluid is preferred. If desired, a separate heating plate can be provided between the back plate and the insulating plate. Each plate of the die-head is provided with feed channels, which are in alignment when the plates are assembled together, through which the heat-plastified polymer is extruded and it has been found that any tendency for the heat-plastified polymer is solidified prematurely in these feed channels with resulting impairment of the extrusion operation or even total blockage of the feed channels can be overcome in some circumstances (depending for example on the size of the feed channels and the extrusion temperature employed) by enlarging the diameter (or cross-sectional area) of the feed channels in the front plate constituting the die-face, or at least the extrusion orifice portions of such feed channels. For example the diameter of the extrusion orifices can be at least 20% greater than that of the feed channels in the back plate and insulating plate.

A convenient way of carrying out the present invention employing a face-cutting technique for manufacturing e.g. expandable polystyrene particles is illustrated in Figure II of the accompanying drawings. Referring to Figure II, polystyrene granules are fed

to the hopper of an extruder 10 which is of the same kind as that described with reference to Figure I except for the die-head 35 which is of a sandwich construction as shown on an enlarged scale in Figure III of the accompanying drawings and will be described in detail later with reference thereto. The pentane-containing polystyrene is extruded from the die-head and immediately face-cut by a two-armed cutter 36 bearing against the face 37 of the die 35, whilst being cooled simultaneously to a temperature between 60°C and 80°C by water droplets sprayed against the die-face 37 from a nozzle 38 supplied with wet steam which condenses to form the water droplets in the vicinity of the die face 37. The resulting particles 39 are carried by the water droplets into slurry tank 40 containing expandable polystyrene particles in suspension in water at 60 to 80°C. A slurry of particles in water at this temperature is continuously pumped from the tank 40 to the base of a normalising tower 23 as described with reference to Figure I; and the normalised particles are removed from the top of the normalising tower 23 and passed via a conveyor 30 to a storage bin 33, the arrangement following the tank 40 being similar to that shown in Figure I to which reference is made in this respect.

The die-head, shown in Figure III, which is the preferred form of a die-head for use in the present invention when face-cutting is used, comprises a body of sandwich construction mounted at the extrusion end of the extruder. The die-head body comprises, in combination, steel front and back plates 41, 42 with a cellulose-filled, phenol formaldehyde resin insulating plate 43 disposed between the front and back plates 41, 42 to provide thermal insulation between the front plate 41, which provides the face portion 37 of the die-head, and the extruder 10. The back plate 42, which is mounted on a flanged portion 10' of the output end of the extruder 10, is provided with a plurality of feed channels 44 for the heat-plastified polymer. The back plate 42 is also provided with a milled channel 45 on its outer face which, when the die-head is assembled and mounted on the extruder 10 with a plate 46 interposed between the back plate 42 and the extruder flanged portion 10', provides a passage through which steam under pressure is passed to maintain the heating plate at about 100°C to 130°C depending on requirements. The plate 46 in addition to closing the channel 45 also provides transverse feed channels 47 in communication through orifices 48 with the feed channels 44 in the back plate 42. The insulating plate 43 has a similar plurality of feed channels 49 each somewhat larger in diameter than the feed channels 44 in the back plate 42 and leading to a similar number of feed channels 50 in the front

plate 41, the feed channels 50 being somewhat larger in diameter than the feed channels 49 in the insulating plate 43 and forming at their open ends 51 the extrusion orifices of the die-head.

The present invention is illustrated by the following Examples:—

EXAMPLE I

Expandable polystyrene particles were manufactured by a strand-cutting method as described above with reference to Figure J, the polystyrene being rendered expandable by the incorporation therein of technical pentane in a 60 millimetre (screw diameter) extruder comprising a heat-plastifying section in which the polystyrene, supplied as granules to the feed hopper of the extruder, was heated to 200°C; and a pentane-injection section maintained at 150°C to which liquid pentane was supplied and homogeneously incorporated in the heat-plastified polystyrene prior to its extrusion into strands at the die-head at a temperature of 110°C. The resulting strands were immediately led into a water bath maintained at 65°C and, after a short residence time therein, the strands were led out of the bath to an adjacent conventional strand cutter by which they were cut into particles 0.06 inch long and 0.02 inch in diameter (average dimensions). The particles immediately fell under gravity into a slurry tank maintained at 65°C from whence a slurry of particles was pumped to the base of a normalising tower in which normalising at 65°C was completed. The period in which the expandable polymer was out of contact with water at 60-80°C, i.e. the time taken for the strand cutting operation, was of the order of 5 seconds. The residence time of the particles in the tower was such that the total normalising period for the particles was 40 minutes. After normalisation the particles were separated from the bulk of the water physically associated therewith and then air-dried.

The resulting expandable polystyrene particles contained on average 5% by weight of pentane and, when subjected to expansion conditions involving contact with live steam at ambient pressure, the particles expanded to form cylindrical beads, the lengths and diameters of which were similar and lay between 0.08 and 0.1 inches; and the beads had a cell size between 70 and 100 microns in diameter. The expanded particles were found to be suitable in all respects for the manufacture of shaped articles e.g. blocks by conventional steam moulding technique.

EXAMPLE II

Expandable polystyrene particles were manufactured by a face-cutting method as described above with reference to Figures II and III, the polystyrene being rendered expandable by the incorporation of technical pentane in the manner described in Example

I except that in order to avoid solidification of polymer in the die-head the temperature of the pentane-containing polystyrene at the die-head was 120°C.

- 5 A sprinkler was disposed adjacent to the front plate for spraying the face of the plate with water droplets at 70°C during operation of the extruder, the water droplets being formed by the condensation of wet steam.
- 10 The cutter was rotated at 30 revolutions per minute and the cut particles were carried down by the droplets of the water spray into a body of water, also at 70°C, and after a residence time therein of 40 minutes to
- 15 achieve normalisation, the particles were separated from the water and cooled to ambient temperature and simultaneously dried in a current of air.

Using polystyrene and operating at the 20 specified extrusion temperature of 120°C, particles of about 1 millimetre in diameter and containing 5% by weight of pentane were obtained which showed no sign of voids. When subjected to expansion conditions involving contact with live steam at ambient pressure the particles expanded to form generally spherical beads having a diameter between 3 and 5 millimetres and a cell size ranging from 70 to 100 microns in diameter which were found suitable in all respects for the manufacture of shaped articles by conventional steam moulding technique.

EXAMPLE III

- Expandable polystyrene particles were manufactured by the face-cutting method the die-head being a multi-orifice die-head from which 32 strands of pentane-containing polystyrene were extruded simultaneously, the die-head comprising a body of sandwich construction having a face portion against which a bladed cutter, rotated during operation at 30 revolutions per minute, was disposed in such a manner as to cut the extrudate into particles at the die face. The die-head body comprised, in combination, steel front and back plates with a cellulose-filled phenol formaldehyde resin insulating plate disposed between the front and back plates to provide heat insulation between the front plate, which formed the face portion of the die-head, and the extruder. The back plate, which was mounted on the extruder at its output end, was provided with 32 feed channels each 1.5 millimetres in diameter. The insulating plate was 1.5 millimetres thick and had 32 feed channels each 3 millimetres in diameter and the front plate, which was 2.5 millimetres thick, also had 32 feed channels of the same diameter as those in the insulating plate, the feed channels in the front plate and in the insulating plate being in register with one another and with those in the back plate. The die-head also embodied a separate heating plate which comprised a copper plate having 32 feed channels each

1.5 millimetres in diameter and was mounted between the insulating plate and the back plate. The heating plate had a thickness of 6 millimetres and was provided with a spiral milling on its outerface which, when the die-head was assembled and mounted on the extruder, provided a 3 millimetre-square channel through which steam was passed to maintain the heating plate at about 100°C.

The extruder employed comprised a 60 millimetre (screw diameter) extruder fitted with a screw having a 20:1 length to diameter ratio and a compression ratio of 3:1 in the heat-plastifying section, the screw being of a design providing a heat-plastifying section in which rapid compression over an initial, one-diameter length of the screw took place; followed by a five-diameter length dispersion section to which technical pentane was supplied for incorporation in the heat-plastified polystyrene. In operation the screw was rotated at 20 revolutions per minute and the temperature conditions in the extruder were such that the temperature profile thereof increased from 80°C at the input end to 100°C at the die-head end, the actual temperature of the pentane-containing heat-plastified polystyrene during extrusion being 110°C.

The face-cutting operation was carried out under a spray of water droplets at 60°C, the cut particles being carried down by the droplets of the water spray into a body of water, also at 60°C, and after a residence time therein of 1 hour to achieve normalisation, the particles were separated from the water and cooled to ambient temperature and simultaneously dried in a current of air. When operating at the specified extrusion temperature of 110°C, particles of about 3 millimetres in diameter and containing 4.8% by weight of pentane were obtained which showed no sign of voids. On expansion with live steam at ambient pressure such expandable polystyrene particles expanded to form generally spherical beads having a diameter of about 1 centimetre and a cell size ranging from 80 to 100 microns in diameter. Such particles were moulded into blocks of cellular polystyrene by conventional steam moulding technique and performed satisfactorily in this application.

WHAT WE CLAIM IS:—

1. A process for the manufacture of expandable particles of a thermoplastic organic polymer (as hereinbefore defined) in which a vaporisable expanding agent is incorporated in a heat-plastified mass of said polymer and the heat-plastified polymer is extruded in filamentary form and cut into particles, which comprises extruding the heat-plastified polymer, containing said expanding agent, into an essentially gaseous environment at atmospheric or substantially atmospheric pressure and immediately cool-

- ing the extruded polymer by contact with a fluid at a temperature between 60 and 80°C in such a manner that the temperature of the extruded polymer does not fall below 60°C.
- 5 maintaining the temperature of the extruded polymer between 60 and 80°C during a normalising period (as herein defined) immediately following extrusion thereof and before and/or after cutting the extruded polymer
- 10 into particles, and thereafter cooling the polymer particles to ambient temperature.
2. A process as claimed in claim 1, which comprises cutting the extruded polymer into particles whilst cooling said extruded polymer by contact with said fluid at a temperature
- 15 between 60 and 80°C.
3. A process as claimed in claim 2, wherein the polymer is extruded from a die-head at which the extruded polymer is face-cut into particles whilst being simultaneously cooled to a temperature between
- 20 60 and 80°C by droplets of a liquid sprayed against the face of said die-head, and the resulting particles are then immediately contacted with a body of said liquid at 60-80°C.
- 25 4. A process as claimed in claim 3, wherein the die-head comprises an outer face plate and an insulating plate providing thermal insulation between said face plate and the body of the extruder to which said die-head is attached.
- 30 5. A process as claimed in claim 4, wherein said die-head comprises a heating plate interposed between said insulating plate and said extruder, said heating plate comprising a passage or passages for the circulation of a fluid at elevated temperature.
- 35 6. A process as claimed in claim 1, which comprises immediately contacting the extruded polymer with a body of fluid at a temperature between 60 and 80°C, cutting the filamentary extruded polymer into particles and substantially immediately contacting the resulting particles with said fluid at
- 40 60-80°C.
7. A process as claimed in claim 6, wherein the polymer is extruded into a body of liquid at 60-80°C from a die-head located adjacent to but spaced from said
- 50 body of liquid and the resulting filamentary extruded polymer is removed from said body of liquid for cutting into particles, which particles are then immediately contacted with a body of liquid at 60-80°C.
- 55 8. A process as claimed in any one of the preceding claims, wherein said thermoplastic organic polymer is polystyrene.
9. A process as claimed in any one of the preceding claims, wherein said vaporisable expanding agent consists of or comprises a- 60 pentane or iso-pentane or a mixture thereof.
10. A process as claimed in any one of the preceding claims, wherein the extrusion temperature of said heat-plastified polymer is between 130 and 160°C and the polymer
- 65 is extruded through orifices below 1.0 millimetre in diameter.
11. A process as claimed in any one of the preceding claims, wherein said fluid or liquid at 60-80°C is water. 70
12. A process as claimed in any one of the preceding claims, wherein at least the major part of the normalisation of the extruded polymer is carried out in a tower containing water maintained at 60 to 80°C, a
- 75 suspension of polymer particles in water at 60 to 80°C being supplied continuously to the base of said tower and said particles being moved progressively up said tower for discharge from the top of said tower. 80
13. A process for the manufacture of expandable particles of a thermoplastic polymer substantially as hereinbefore described with reference to Figure I of the accompanying drawings. 85
14. A process for the manufacture of expandable polystyrene particles substantially as hereinbefore described with reference to Example I.
15. A process for the manufacture of expandable particles of a thermoplastic polymer substantially as hereinbefore described with reference to Figures II and III of the accompanying drawings. 90
16. A process for the manufacture of expandable polystyrene particles substantially as hereinbefore described with reference to Example II or Example III. 95
17. Expandable particles of a thermoplastic polymer when manufactured by the process claimed in any one of the preceding claims. 100
18. Expanded particles of a thermoplastic polymer obtained by subjecting to expansion conditions expandable particles as claimed in claim 17. 105
19. A shaped article manufactured by moulding expanded particles as claimed in claim 18.

WILLENS & ROBBINS
Chartered Patent Agents
Shell Centre
London, S.E.1.
Agents for the Applicants.

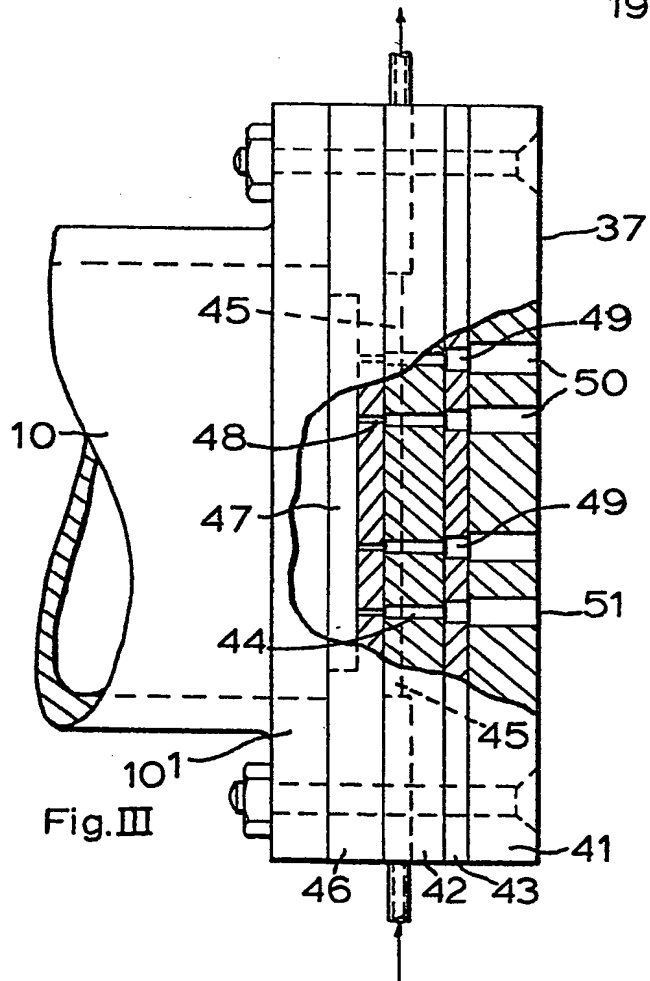
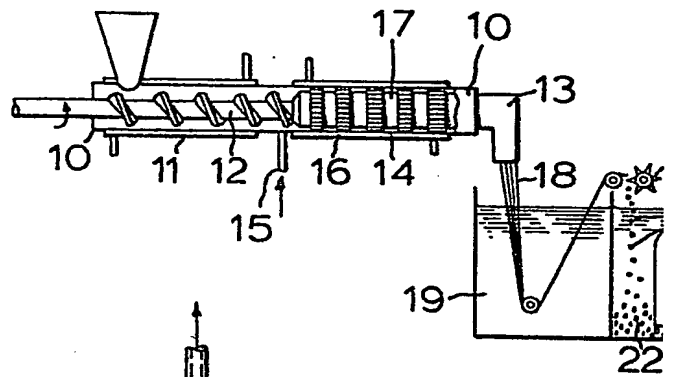


Fig. III

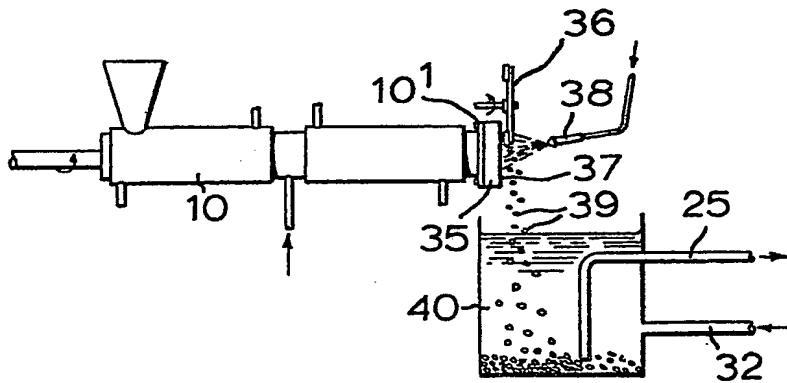
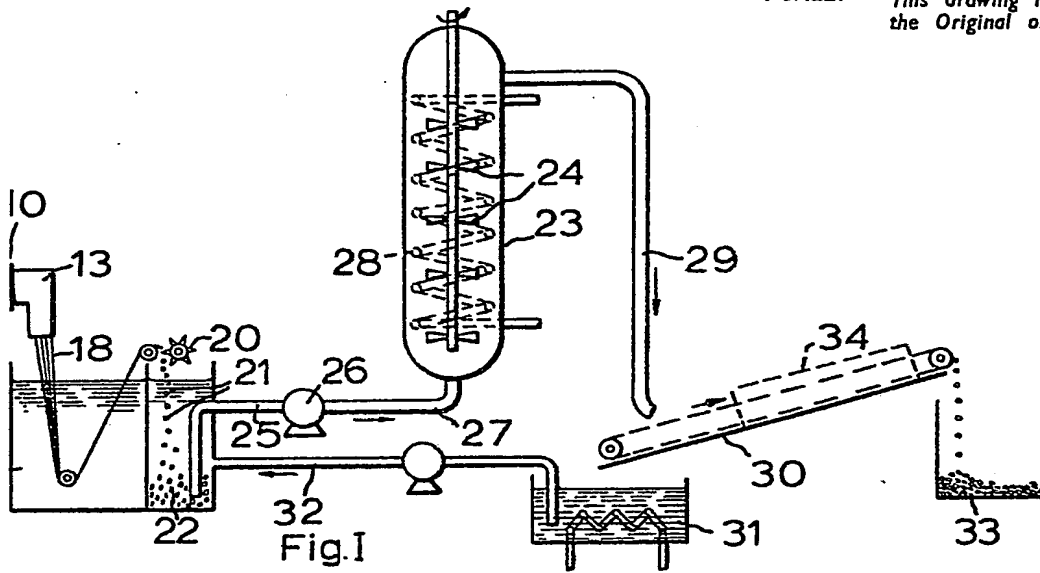


Fig. 1

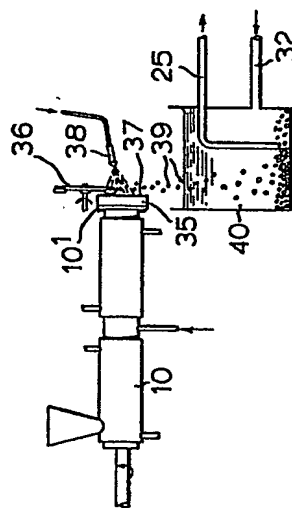


Fig. II

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☒ FADED TEXT OR DRAWING
- ☒ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☐ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.